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13. ABSTRACT (Maximum 200 words)
This report describes the characterization and thermal stability of nanocrystalline oxide composites for thermal barrier coating applications. Electrophoretic deposition was used to coat the nanocomposite powders onto nickel substrates. Prior to electrophoretic deposition of the nanocomposite powders, bond coats were applied to the nickel substrates by either plasma spraying or by electrophoretic deposition. Two types of coatings (double-layered and compositionally graded coatings) were deposited, characterized and tested for thermal stability. Oxidation studies on the Al₂O₃-Y₂O₃-ZrO₂ nanocomposite coatings were performed to better understand the mechanism of failure in the thermal barrier coatings.

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Processing and Deposition of Nanocrystalline Oxide Composites for
Thermal Barrier Coatings

Technical Report on ONR Grant No. N00014-95-1-0626
for the Period of October 1, 2000-December 31, 2000

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1. Plasma Spraying of CoNiCrAlY Bond Coat and Electrophoretic Deposition of Al_2O_3 - Y_2O_3 - ZrO_2 Nanocomposites

As described in a previous report [1], electrophoretic deposition (EPD) was used as the method for depositing thermal barrier coatings (TBC). In this report, we used the conventional technique of plasma spraying to deposit a CoNiCrAlY bond coat on the substrates. Following thermal spraying of the bond coat, EPD was used to deposit consecutive layers of Al_2O_3 and Al_2O_3 -YSZ coatings as well as to deposit a compositionally graded coating of YSZ and Al_2O_3 . The compositional gradient was confirmed using energy dispersive X-ray analysis (EDX). We examined the thermal stability of the double-layered coatings and graded coatings. In addition, substrate oxidation was characterized using X-ray diffraction (XRD) to monitor oxidation of the underlying Ni substrate.

The CoNiCrAlY bond coats were plasma sprayed onto the Ni substrates by PERMA. The thickness of the plasma sprayed bond coats was $\sim 50 \mu\text{m}$. The bond coats were pretreated at 600°C in air for 1 hr prior to electrophoretic deposition of the other coatings. For electrophoretic deposition of the Al_2O_3 layer, a coating solution consisting of ethanol, water, nitric acid, and Al_2O_3 powder was used. The solution composition was similar to that reported previously [1]. Al_2O_3 powder was first mechanically milled for 24 hr to decrease particle size and then added to the coating solution to form a slurry. The slurry was then ultrasonicated for 15 min and aged for 24 hr. Ni electrodes were then lowered into the slurry and a DC voltage ($\sim 40 \text{ V/cm}$) was applied for 60 sec, resulting in a $\sim 20 \mu\text{m}$ -thick Al_2O_3 coating. The electrodes were then removed from the slurry, and the coated substrate was dried at 110°C for 2 hr. Following drying, the Al_2O_3 coatings were cold isostatically pressed at 50,000 psi to increase the mechanical stability. For the subsequent deposition of a $50 \mu\text{m}$ -thick Al_2O_3 -YSZ layer, a coating solution containing 5 wt% Al_2O_3 -1.2 wt% Y_2O_3 -93.8 wt% ZrO_2 was used. Following drying at 110°C for 2 hr, the double-layered coatings were cold isostatically pressed at 50,000 psi to increase the mechanical stability.

For the compositionally graded coating, deposition began with a pure Al_2O_3 suspension. A stream of YSZ suspension, consisting of 1.3 wt% Y_2O_3 -98.7 wt% ZrO_2 was slowly and continuously injected into the well-stirred Al_2O_3 bath. The injection rate of the YSZ suspension controlled the relative content of YSZ in the Al_2O_3 suspension. A DC voltage (~ 40 V/cm) was applied for 300 sec, resulting in a ~ 60 μm -thick Al_2O_3 - Y_2O_3 - ZrO_2 graded coating.

2. Compositional Analysis of the Al_2O_3 - Y_2O_3 - ZrO_2 Nanocomposite Coatings

Figure 1 shows an optical micrograph of the double-layered $\text{Al}_2\text{O}_3/\text{Al}_2\text{O}_3$ -YSZ coatings. The micrograph shows a discrete, ~ 20 μm -thick Al_2O_3 interlayer between the bond coat and Al_2O_3 -YSZ coatings. The Al_2O_3 interlayer may provide a barrier to prevent oxygen diffusion, suppressing bond coat oxidation and subsequent thermal barrier coating failure.

Scanning electron microscopy equipped with energy dispersive X-ray analysis was used to measure Al, Zr, and Ni contents in the graded Al_2O_3 -YSZ thermal barrier coating. Figure 2 shows the relative at% of Al, Zr and Ni as a function of coating thickness. The region from 0 to ~ 40 μm from the substrate represents the plasma sprayed bond coat (CoNiCrAlY). Al content begins to increase at ~ 40 μm from the Ni substrate surface, where deposition of the Al_2O_3 coating begins. The region from ~ 40 to ~ 60 μm consists of pure Al_2O_3 coating, prior to YSZ injection into the EPD bath. At a distance of ~ 60 microns, injection of the YSZ suspension into the Al_2O_3 bath results in a gradual decrease in Al content from the substrate, a steady increase in Zr content to give a graded coating. At ~ 80 - 100 μm from the substrate, the relative Al content dropped to 0, indicating a pure YSZ coating. This analysis confirms that a smooth gradient in Al_2O_3 and YSZ composition has been achieved in the coating. The graded coating could prove valuable for thermal barrier coating applications, providing a smooth change in thermal and mechanical properties across the coating.

3. Thermal Stability of Al_2O_3 - Y_2O_3 - ZrO_2 Nanocomposite Coatings

The double-layered $\text{Al}_2\text{O}_3/\text{Al}_2\text{O}_3$ -YSZ coatings and the graded Al_2O_3 -YSZ coatings were tested for thermal stability. The coated Ni substrates were pretreated in argon at 1150 $^\circ\text{C}$ for 1 hr (ramp = 10 $^\circ\text{C}/\text{min}$). They were then tested for long-term thermal stability by heating to 1150 $^\circ\text{C}$ in air (ramp = 10 $^\circ\text{C}/\text{min}$) for 0–5 hr. The samples were inspected visually every hour for coating spallation and Ni substrate oxidation. All experiments were repeated three times.

The graded Al_2O_3 -YSZ coatings lasted 3 hr following heat treatment at 1150 $^\circ\text{C}$ in air before visible spallation occurred. Al_2O_3 -YSZ coatings prepared without the compositional gradient lasted up to 14 hr before visible spallation was observed [2]. The high alumina content in the graded coating may cause stress due to a mismatch in the thermal expansion coefficient between Al_2O_3 and YSZ within the initial layer. Prior

results have shown a negative effect of Al_2O_3 with contents ≥ 20 wt% [2]. Thermal stability of the graded system may be further improved by lowering the Al_2O_3 content < 20 wt%.

The double-layered $\text{Al}_2\text{O}_3/\text{Al}_2\text{O}_3\text{-YSZ}$ thermal barrier coatings were also stable up to 3 hr at 1150°C in air before visible spallation was observed. The Al_2O_3 interlayer may have also lowered the mechanical stability of the TBC. An alternative coating procedure for the Al_2O_3 interlayer, such as plasma spraying or spray coating, may improve the mechanical and thermal stability of the double-layered thermal barrier coating.

4. Oxidation of Bond Coat and Ni Substrate

Previous results suggest that the optimum coating consists of a plasma-sprayed bond coat, followed by an electrophoretically deposited thermal barrier coating consisting of 5 wt% Al_2O_3 -1.2 wt% Y_2O_3 -93.8 wt% ZrO_2 [2]. The optimized samples were heat treated at 1150°C in air for 0–10 hr (Figure 3(a)-(c)). Following the heat treatment, the samples were examined for bond coat and Ni substrate oxidation using XRD. A relatively small NiO peak appeared after 10 hr of heat treatment at 1150°C (Figure 3(c)), which corresponded well with the observed coating spallation after 14 hr of heat treatment [2]. Oxidation of the bond coat and the Ni substrate appeared to be the primary cause of failure for thermal barrier coatings deposited electrophoretically.

For comparison, a sample prepared with electrophoretically deposited bond coat and 5 wt% Al_2O_3 -1.2 wt% Y_2O_3 -93.8 wt% ZrO_2 heat treated for 10 hr was also examined for substrate oxidation (Figure 3(d)). The XRD pattern of the sample prepared with EPD bond coat shows a large NiO peak after 10 hr of heat treatment, which corresponded with the observed coating spallation after only 6 hr of heat treatment [2]. The EPD bond coat proved less effective in oxidation resistance compared with the plasma-sprayed bond coat.

5. Future Work

Future work will focus on an alternative coating route for applying the optimized Al_2O_3 -YSZ composition. Al_2O_3 -YSZ coating solution will be prepared by mixing the optimized Al_2O_3 -YSZ powder with an Al_2O_3 gel, forming a viscous slurry. The powder/gel ratio in the slurry will be optimized for thermal and mechanical stability. This alternative coating procedure may potentially yield thick ($\sim 500\ \mu\text{m}$) coatings not attainable by electrophoretic deposition. The coatings will be applied to uncoated and bond-coated Ni substrates, and tested for thermal stability.

6. References

- [1] Ying, J.Y., Technical Report on ONR Grant No. N00014-95-1-0626 for the period of October 1, 1999-December 31, 1999.
- [2] Ying, J.Y., Technical Report on ONR Grant No. N00014-95-1-0626 for the period of July 1, 2000-September 30, 2000.

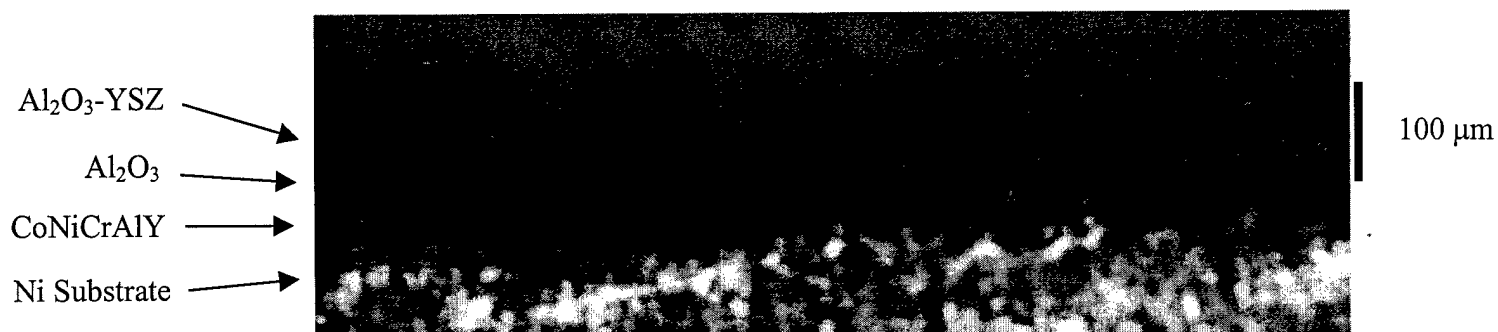


Figure 1. Optical micrograph of the electrophoretically deposited $\text{Al}_2\text{O}_3/\text{Al}_2\text{O}_3\text{-YSZ}$ double-layered coatings.

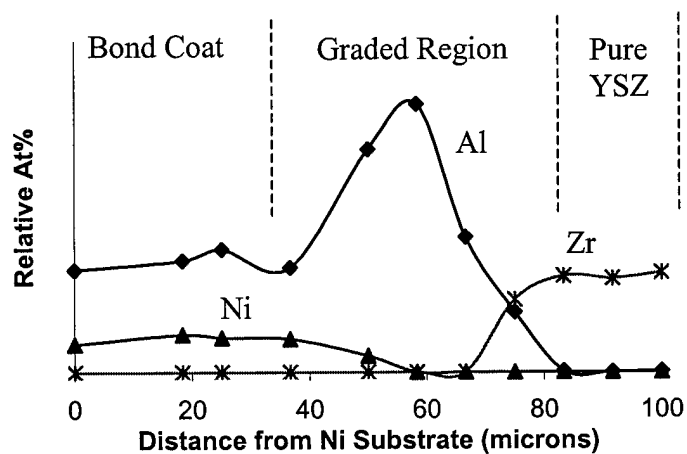


Figure 2. Relative at% of Al, Zr and Ni as a function of coating thickness for the graded $\text{Al}_2\text{O}_3\text{-YSZ}$ coating. The relative Zr, Al and Ni compositions were obtained with scanning electron microscopy equipped with energy dispersive X-ray analysis.

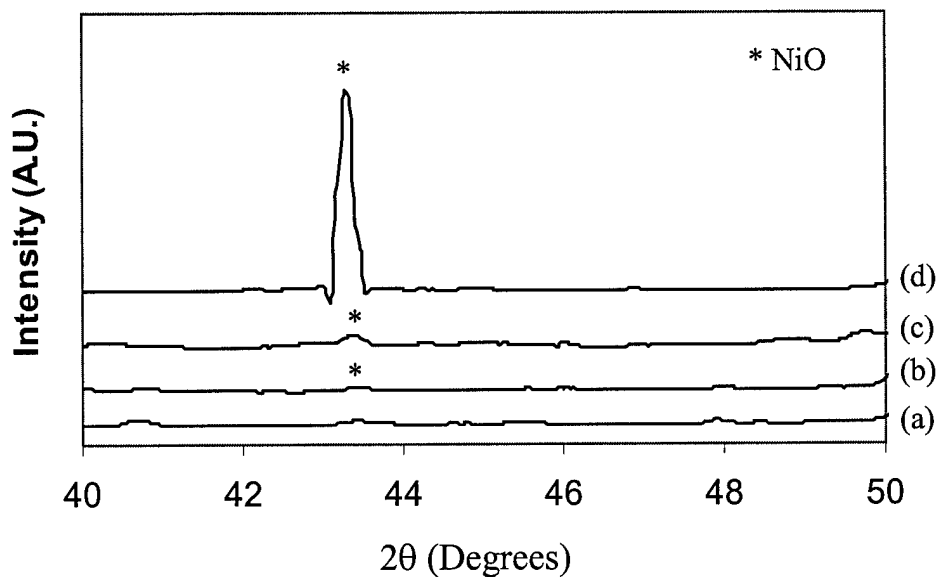


Figure 3. X-ray diffraction patterns of 5 wt% Al_2O_3 -1.2 wt% Y_2O_3 -93.8 wt% ZrO_2 TBC electrophoretically deposited on a Ni substrate with plasma-sprayed CoNiCrAlY bond coat, following (a) 0, (b) 4, and (c) 10 hours of heat treatment at 1150 °C in air. Sample (d) was heat treated for 10 hours and consisted of an electrophoretically deposited bond coat.